LUUS P U88U2

37

(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: **0 350 941 A2**

(12) **EUROPEAN PATENT APPLICATION**

(21) Application number: 89112941.3

(51) Int. Cl.⁴: H01L 41/08 , H01L 41/22

(22) Date of filing: 14.07.89

(30) Priority: 15.07.88 JP 175050/88

(43) Date of publication of application:
17.01.90 Bulletin 90/03

(84) Designated Contracting States:
DE FR GB

(71) Applicant: HITACHI, LTD.
6, Kanda Surugadai 4-chome Chiyoda-ku
Tokyo(JP)

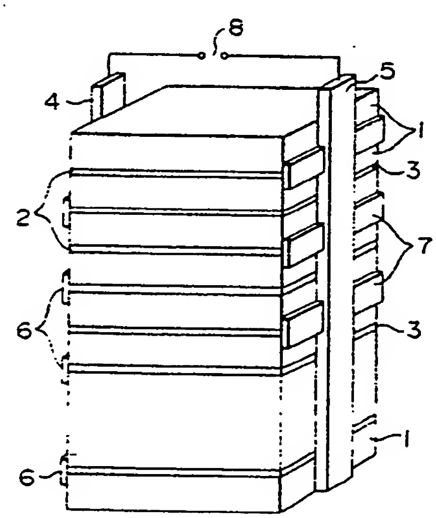
(72) Inventor: Kohno, Akiomi
2625-242, Koibuchi Tomobemachi
Nishiibaraki-gun Ibaraki-ken(JP)
Inventor: Arai, Masatugu
Niihariryo, 3602 Shimoinayoshi Chiyodamura
Niihari-gun Ibaraki-ken(JP)
Inventor: Harada, Takeshi
5200-1-306, Terada
Toride-shi(JP)

(74) Representative: Patentanwälte Beetz sen. -
Beetz jun. Timpe - Siegfried -
Schmitt-Fumian
Steinsdorfstrasse 10
D-8000 München 22(DE)

(54) **Stack-type piezoelectric element, process for producing the same, and stack-type piezoelectric device.**

(57) According to the present invention, there are provided a stack-type piezoelectric element constructed of a stacked body comprising plural layers (1) of sintered piezoelectric ceramic material and electrodes (2, 3) which lie between the respective layers (1, 1) of the piezoelectric ceramic material, wherein said electrodes (2, 3) are formed of a material capable of diffusion-bonding with said piezoelectric ceramic material at a lower temperature than the sintering temperature of said ceramic material, a process for producing the same, and a stack-type piezoelectric device using said stack-type piezoelectric element.

FIG. 1



EP 0 350 941 A2

STACK-TYPE PIEZOELECTRIC ELEMENT, PROCESS FOR PRODUCING THE SAME, AND STACK-TYPE PIEZOELECTRIC DEVICE

BACKGROUND OF THE INVENTION

The present invention relates to a stack-type piezoelectric element, a process for producing the same, and a device using said element. In particular, it relates to a stack-type piezoelectric element having a high reliability as a piezoelectric actuator, a process for producing the same, and also to a device using said element.

Since a piezoelectric ceramic generates an electric voltage when a force is applied thereto and produces a displacement or a force when voltage is applied thereto, it is highly useful for use as actuators or sensors. When a piezoelectric ceramic is applied to actuators or sensors, it is usually used in the form of a stacked body thereof with electrodes. Such a stacked body is called a stack-type piezoelectric element.

Stack-type piezoelectric elements are produced, like other ceramic laminates, generally by the green sheet method. A piezoelectric ceramic green sheet is a sheet-formed material obtained by calcining the starting material for piezoelectric ceramic, adding to the resulting powder a suitable binder etc. to form a slurry, and then molding the slurry into a sheet. The green sheet thus obtained is printed with a metal paste by means of screen printing or like methods to form an electrode part. A desired number of thus prepared sheets are stacked, dried, and then sintered at one thousand several hundred degrees (Centigrade) to be converted into ceramic. The product thus obtained is the piezoelectric element stacked body of the prior art. For use as the final product, the piezoelectric device is further subjected to lead formation and resin coating and polarization treatment by application of voltage. Such a process for producing stacked bodies using conventional green sheet method is disclosed, for example, in Japanese Patent Application Kokai (Laid-open) Nos. 60-41273 (1985), 60-91800 (1985) and 60-229380 (1985). A process flow diagram for prior stack-type piezoelectric device is shown in Fig. 3.

In the above-mentioned prior process shown in Fig. 3 the sintering of green sheets and the baking of electrodes are carried out simultaneously, so that it is seemingly a simple process because of relatively small man-hour required. However, the binder etc. present in green sheets are difficultly removed by heating owing to obstruction by electrode layers and, if an additional step of removing the binder by heating is taken, the ceramic body is apt to undergo cracking, and destacking deformation.

Furthermore, since the stacked body of green sheets and electrodes is sintered at a high temperature and at one time, and since the contraction of ceramic during sintering is large, a large strain (in other words, internal stress) develops inside the piezoelectric ceramic stacked body. Accordingly, when a high voltage is applied to the stacked body or when it is used repeatedly for a long period the stacked body undergoes mechanical breakage.

Moreover, a large strain develops also in the polarization treatment conducted after sintering, which also leads to lowering or reliability of strength.

Thus, the stack-type piezoelectric element of the prior art is itself poor in reliability of strength and hence not fully satisfactory for use as actuators etc.

Further, the electrode provided between piezoelectric ceramics has been conventionally made of silver of silver-palladium (Ag-Pd) alloy. This is because, since the sintering of green sheets of piezoelectric ceramic and the stacking thereof with electrodes are simultaneously effected at about 1300°C, a noble metal stable at high temperature must be used as the electrode material. However, this gives rise to the following problem. That is, water can penetrate into the element through molding resin protecting the side face of the piezoelectric element, pass through between electrodes, and further form an aqueous layer between the stacked body and the resin. Then, silver is dissolved out as ions (Ag^+), which are then attracted by an electric field to accumulate in the vicinity of neighboring electrode, forming a current-carrying path. In consequence, short circuit of electrodes occurs in the prior art piezoelectric element and device when it is operated in a humid atmosphere. The above-mentioned phenomenon of current-carrying path formation is generally called "migration" and is a serious problem common to electrical parts using electrodes made of silver or silver alloy.

SUMMARY OF THE INVENTION

The stack-type piezoelectric element of the present invention is constructed of a stacked body comprising plural layers of sintered piezoelectric ceramic material and electrodes which lie between the respective layers of the piezoelectric ceramic material, wherein said electrodes are formed of a material capable of diffusion-bonding with said ceramic material at a lower temperature than the sintering temperature of the piezoelectric ceramic

material. Examples of the most suitable materials for the electrodes include metallic materials comprising one or more layers of at least one member selected from the group consisting of aluminum and aluminum alloys or one or more layers of at least one member selected from the group consisting of nickel and nickel alloys.

The stack-type piezoelectric device (e.g. actuator and sensor) of the present invention comprises a stacked body formed of plural layers of sintered piezoelectric ceramic material and electrodes which lie between the respective layers of the piezoelectric ceramic material, said respective electrodes being provided such that the positive poles and the negative poles are alternately arranged in the stacking direction, an external lead which connects the positive poles together, and an external lead which connects the negative poles together, both provided at the side of said stacked body, wherein said stacked body is the above-mentioned stack-type piezoelectric element of the present invention.

The process for producing a stack-type piezoelectric element of the present invention comprises stacking sintered piezoelectric ceramic material layers and electrode layers alternately, heating, or heating under applied pressure, the stacked product at a temperature lower than the sintering temperature of the piezoelectric ceramic material and keeping it at the temperature for a predetermined time, thereby to effect bonding, preferably diffusion bonding.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a perspective view of a stack-type piezoelectric device of one embodiment of the present invention.

Fig. 2 is a perspective view of an electrode layer used in another embodiment of the present invention.

Fig. 3 is a process flow diagram for producing a stack-type piezoelectric device and here the main description of the invention is enclosed by a broken line.

DETAILED DESCRIPTION OF THE INVENTION

The "sintered piezoelectric ceramic" referred to in the invention is the product obtained by sintering a piezoelectric ceramic green sheet.

The piezoelectric element according to the present invention is preferably cooled after the bonding step without removal of applied pressure. Particularly in the cooling process conducted after the bonding step, the piezoelectric ceramic is desirably subjected to a polarization treatment by applying a voltage thereto at a higher temperature by 20-70 °C than the Curie point of the ceramic. The "Curie point" referred to herein is a temperature at which the piezoelectric ceramic loses its characteristic property, and the "polarization treatment" is a step of treatment of ceramic for converting it into a piezoelectric device.

In general, a piezoelectric ceramic material is a ferromagnetic material having a perovskite-type crystal structure. At a higher temperature than the Curie point the ceramic material exists as a crystalline phase of the cubic system, but at a lower temperature than the Curie point it assumes a crystalline phase deformed from the cubic system, which results in development of dislocation, in other words polarity (or orientation), in crystal structure. A piezoelectric ceramic immediately after sintering is an aggregate of crystal grains and the respective crystal grain comprises parts of different polarities mingled with one another. In a piezoelectric ceramic which has been subjected to sintering alone, the sum of polarity vectors is zero for respective crystal grains and the ceramic is nonpolar as a whole. When a predetermined electric field is applied thereto, the direction of polarity is determined for respective crystal grains. This phenomenon is termed polarization and such a treatment is termed the polarization treatment.

The electrode material may be used in any of the forms of film, foil and paste. Preferred conventional materials are pure aluminum, aluminum alloys, nickel and nickel alloys. When an aluminum electrode is used, it is preferably of a three-layer structure wherein the both skin materials are aluminum-silicon alloy and the core material is aluminum. The material employed for a nickel electrode is nickel used alone or a nickel alloy or, particularly preferably, a solid solution of nickel with either chromium, phosphorus, or a compound thereof. A particularly preferable nickel electrode material has a structure comprising a nickel core material and nickel alloy skin materials formed on the both sides thereof. When aluminum alloy of a three-layer structure is used as the electrode material, the diffusion bonding temperature is 580 - 660 °C. In forming a stacked body by application of heat and pressure, at a diffusion bonding temperature of 580 -660 °C, only the skin layers of the electrode become liquid, so that the oxide film on the aluminum alloy electrode surface is broken and the liquid phase of the active aluminum alloy acts to enhance the bonding reaction with the piezoelectric ceramic. On the other hand, aluminum of the core material remains solid and keeps a fixed electrode thickness. In the case of nickel alloy of a three-layer structure, the diffusion bonding temperature is 875 - 1200 °C.

The stacking form is not restricted to a planar

Claims

1. A stack-type piezoelectric element constructed of a stacked body comprising plural layers (1) of sintered piezoelectric ceramic material and electrodes (2,3) which lie between the respective layers (1,1) of the piezoelectric ceramic material, wherein said electrodes (2,3) are formed of a material capable of diffusion-bonding with said ceramic material at a lower temperature than the sintering temperature of the piezoelectric ceramic material.

2. A stack-type piezoelectric element constructed of a stacked body comprising plural layers (1) of sintered piezoelectric ceramic material and electrodes (2,3) which lie between the respective layers (1,1) of the piezoelectric ceramic material, wherein said respective electrodes are formed of a metallic material comprising one or more layers (22,21) of at least one member selected from aluminum and aluminum alloy or one or more layers of at least one member selected from nickel and nickel alloy.

3. A stack-type piezoelectric device comprising a stacked body formed of plural layers (1) of sintered piezoelectric ceramic material and electrodes (2,3) which lie between the respective layers (1,1) or the piezoelectric ceramic material, said respective electrodes (2,3) being provided such that the positive poles and the negative poles are alternately arranged in the stacking direction, an external lead (4) which connects the positive poles together, and an external lead (5) which connects the negative poles together, both provided at the side of said stacked body, wherein said stacked body is a stacktype piezoelectric element according to claim 1.

4. A stack-type piezoelectric device comprising a stacked body formed of plural layers (1) of sintered piezoelectric ceramic material and electrodes (2,3) which lie between the respective layers (1,1) of the piezoelectric ceramic material, said respective electrodes (2,3) being provided such that the positive poles and the negative poles are alternately arranged in the stacking direction, an external lead (4) which connects the positive poles together, and an external lead (5) which connects the negative poles together, both provided at the side of said stacked body, wherein said stacked body is a stack-type piezoelectric element according to claim 2.

5. A process for producing a stack-type piezoelectric element which comprises stacking sintered piezoelectric ceramic material layers (1) and electrode layers (2,3) alternately, and then heating the stacked product at a lower temperature than the sintering temperature of said piezoelectric ceramic material to effect bonding.

6. A process for producing a stack-type piezoelectric element according to claim 5 which further comprises applying a voltage to the piezoelectric ceramic at a higher temperature by 20-70°C than the Curie point of said ceramic in the course of cooling succeeding to the bonding step, thereby to effect polarization of said ceramic.

7. A process for producing a stack-type piezoelectric element which comprises stacking sintered piezoelectric ceramic layers (1) and electrode layers (2,3) alternately, and then applying heat and pressure to the stacked product at a lower temperature than the sintering temperature of said piezoelectric ceramic material to effect bonding.

8. A process for producing a stack-type piezoelectric element according to claim 7 wherein the stacked product is cooled without removal of applied pressure after the bonding step.

9. A process for producing a stack-type piezoelectric element according to claim 7 which further comprises applying a voltage to the piezoelectric ceramic at a higher temperature by 20-70°C than the Curie point of said ceramic in the course of cooling succeeding to the bonding step, thereby to effect polarization of said ceramic.

10. A process for producing a stack-type piezoelectric element which comprises sintering piezoelectric ceramic green sheets, then providing electrode layers (2,3) between the respective ceramic layers (1), and heating the whole at a lower temperature than that in said sintering, thereby diffusion-bonding the ceramic materials with the electrode materials.

11. A process for producing a stack-type piezoelectric element according to claim 10 which further comprises applying a voltage to the piezoelectric ceramic at a higher temperature by 20-70°C than the Curie point of said ceramic in the course of cooling succeeding to the bonding step, thereby to effect polarization of said ceramic.

FIG. 1

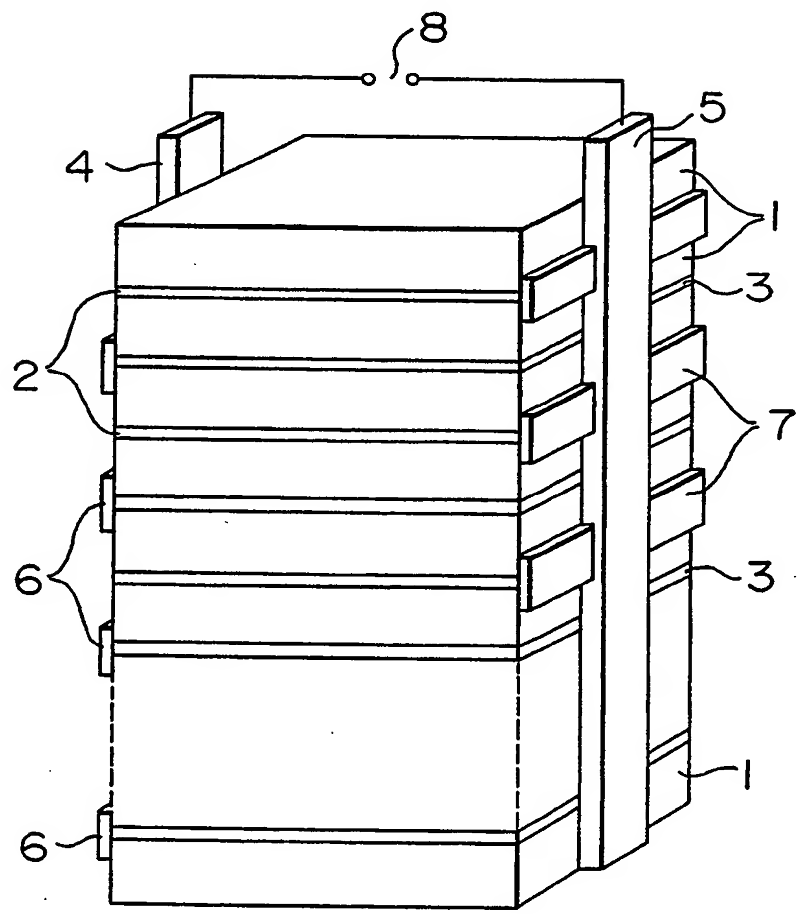
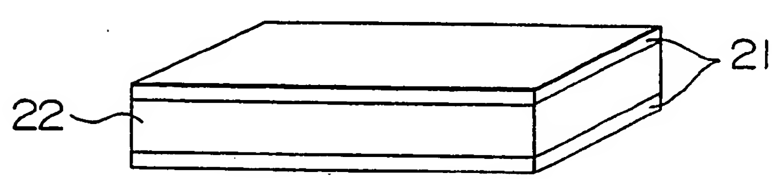
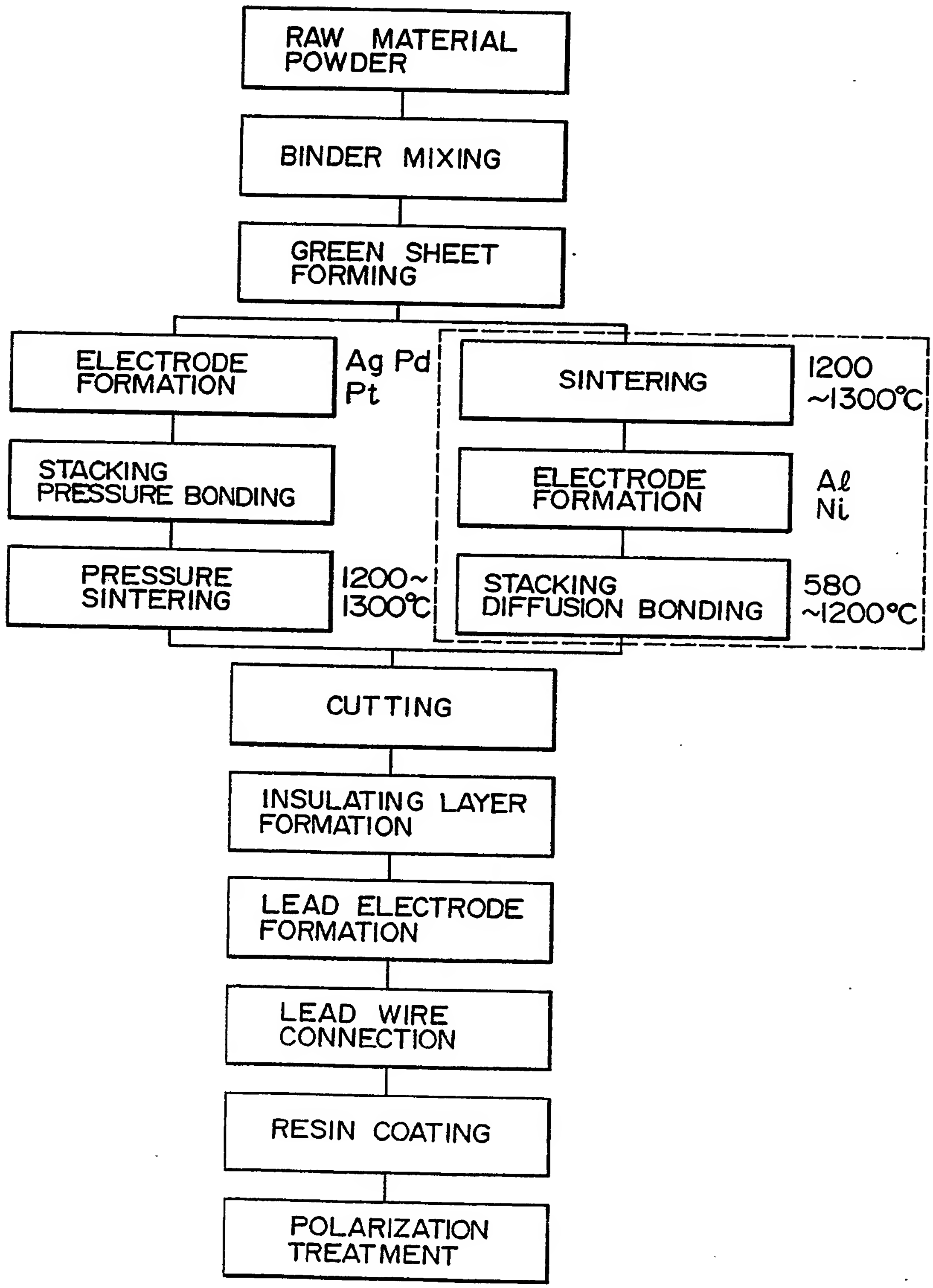


FIG. 2



F I G. 3


RAW MATERIAL POWDER
BINDER MIXING
GREEN SHEET FORMING
ELECTRODE FORMATION
Ag Pd
Pt
STACKING PRESSURE BONDING
PRESSURE SINTERING
1200~
1300℃
SINTERING
1200
~1300℃
ELECTRODE FORMATION
Aℓ
Ni
STACKING DIFFUSION BONDING
580
~1200℃
CUTTING
INSULATING LAYER FORMATION
LEAD ELECTRODE FORMATION
LEAD WIRE CONNECTION
RESIN COATING
POLARIZATION TREATMENT

(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: **0 350 941 A3**

(12) EUROPEAN PATENT APPLICATION

(21) Application number: 89112941.3

(22) Date of filing: 14.07.89

(30) Priority: 15.07.88 JP 175050/88

(43) Date of publication of application: 17.01.90 Bulletin 90/03

(84) Designated Contracting States: DE FR GB

(88) Date of deferred publication of the search report: 27.12.90 Bulletin 90/52

(51) Int. Cl.5: H01L 41/08, H01L 41/22

(71) Applicant: HITACHI, LTD.
6, Kanda Surugadai 4-chome
Chiyoda-ku, Tokyo 100(JP)

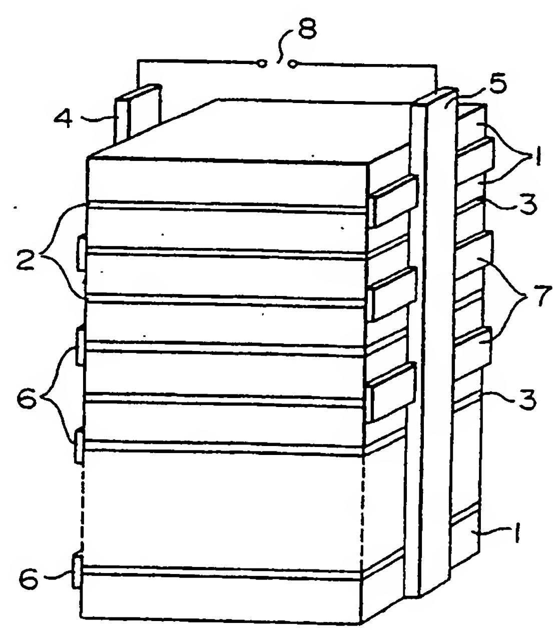
(72) Inventor: Kohno, Akiomi
2625-242, Koibuchi Tomobemachi
Nishiibaraki-gun Ibaraki-ken(JP)
Inventor: Arai, Masatugu
Niihariryo, 3602 Shimoinayoshi Chiyodamura
Niihari-gun Ibaraki-ken(JP)
Inventor: Harada, Takeshi
5200-1-306, Terada
Toride-shi(JP)

(74) Representative: Patentanwälte Beetz sen. - Beetz jun. Timpe - Siegfried - Schmitt-Fumian
Steinsdorfstrasse 10
D-8000 München 22(DE)

(54) Stack-type piezoelectric element, process for producing the same, and stack-type piezoelectric device.

(57) According to the present invention, there are provided a stack-type piezoelectric element constructed of a stacked body comprising plural layers (1) of sintered piezoelectric ceramic material and electrodes (2, 3) which lie between the respective layers (1, 1) of the piezoelectric ceramic material, wherein said electrodes (2, 3) are formed of a material capable of diffusion-bonding with said piezoelectric ceramic material at a lower temperature than the sintering temperature of said ceramic material, a process for producing the same, and a stack-type piezoelectric device using said stack-type piezoelectric element.

FIG. 1



EP 0 350 941 A3

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 89 11 2941

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	EP-A-0 094 078 (NEC) * page 27, lines 4-9; page 29, line 13 - page 30, line 16; page 31, lines 1-6 * -----	1-11	H 01 L 41/08 H 01 L 41/22
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			H 01 L

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	27-09-1990	JUHL A.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document